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Applicant WILLIMANN, Hongli et al	

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L3 ANSWER 1 OF 1 JAPIO COPYRIGHT 2002 JPO
ACCESSION NUMBER: 1988-099285 JAPIO
TITLE: WATER AND OIL REPELLENT
INVENTOR: OMORI AKIRA; INUKAI HIROSHI
PATENT ASSIGNEE(S): DAIKIN IND LTD
PATENT INFORMATION:

PATENT NO	KIND	DATE	ERA	MAIN IPC

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INT. PATENT CLASSIF.:

MAIN: C09K003-18
SECONDARY: C08F020-22; C08F020-22

ABSTRACT:

PURPOSE: To provide a water and oil repellent capable of forming film which is uniform and tough and high in the adhesiveness to articles treated therewith, consisting of a fluorine-contg. polymer containing specified quantities of specific fluorine-contg. acrylate.
CONSTITUTION: The objective water and oil repellent consisting of a fluorine- contg. polymer containing $\geq 10\text{wt}\%$ of a fluorine-contg. acrylate of formula [X is F, Cl or -CFX<SP>1</SP>X<SP>2</SP> (X<SP>1</SP> and X<SP>2</SP> are each H or F); Y is $1\sim 3\text{C}$ alkylene, -CH<SB>2</SB>CH<SB>2</SB>N(R)SO<SB>2</SB>- (R is $1\sim 4\text{C}$ alkyl), or -CH<SB>2</SB>CH(OZ)CH<SB>2</SB>- (Z is H or acetyl); Rf is $3\sim 21\text{C}$ fluoroalkyl or $3\sim 21\text{C}$ fluoroalkyl containing O<SB>1</SB> \sim 10</SB> in the carbon chain (but any of the Os are not mutually adjacent)]. Said polymer can be prepared by radical or anionic polymerization.
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昭63-99285

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⑭ 発明の名称 撥水撥油剤

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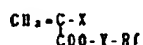
明 細 書

1. 発明の名称

撥 水 撥 油 剤

2. 特許請求の範囲

1. 式:



(式中、X はフッ素原子、塩素原子または
-CPX'X² 基(但し、X' および X² は同一
または相異なり水素原子またはフッ素原子
である。)、Y は炭素原子数1~3のアル
キレン基、-CH₂CH₂N(R)SO₂- 基(但し、R
は炭素原子数1~4のアルキル基である。)
または-CH₂CH(OR)CH₂- 基(但し、Z は水素
原子またはアセチル基である。)、Rf は炭
素原子数3~21のフルオロアルキル基また
は炭素原子数中に1~10の酸素原子を含む
炭素原子数3~21のフルオロアルキル基
(但し、酸素原子同士が隣接することはな
い。)を示す。)

で置換される含フッ素アクリレートを少なく

とも10重量%含む含フッ素重合体からなる撥
水撥油剤。

3. 発明の詳細な説明

(産業上の利用分野)

本発明は、含フッ素撥水撥油剤に関する。

(従来の技術)

フルオロアルキルメタアクリレート重合体等の
含フッ素重合体が撥水撥油剤として使用できるこ
とは公知である(例えば、特公昭47-40467号公
報参照)。

しかし、従来公知の撥水撥油性を有する重合体
は、被処理物品に対してもなじみが悪くまた膜強
度も小さいため、少し擦ったりすると簡単に剥が
れてしまうという問題を有している。

(発明の目的)

本発明者らは、種々の含フッ素アクリレート重
合体を作り、その造膜性、被処理物品に対する撥
水性、膜強度等を調べたところ、特定の含フッ素
アクリレートを構成成分とする重合体がこれら性
質に優れていることを見出し、本発明に達したも

のである。

本発明の目的は、均一かつ強靱で被処理物品に対する接着性が良好な皮膜を形成することができる含フッ素撥水撥油剤を提供することである。

(発明の構成)

本発明は、式：



(式中、X はフッ素原子、塩素原子または $-\text{CFX}'\text{X}''$ 基 (但し、X' および X'' は同一または相異なり水素原子またはフッ素原子である。)、Y は炭素原子数 1~3 のアルキレン基、 $-\text{CH}_2\text{CH}_2\text{N(R)}\text{SO}_2$ -基 (但し、R は炭素原子数 1~4 のアルキル基である。) または $-\text{CH}_2\text{CH}(\text{OZ})\text{CH}_2$ -基 (但し、Z は水素原子またはアセチル基である。)、R' は炭素原子数 3~21 のフルオロアルキル基または炭素原子数中に 1~10 の酸素原子を含む炭素原子数 3~21 のフルオロアルキル基 (但し、酸素原子同士が隣接することはない。) を示す。)

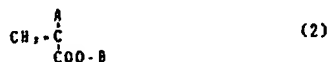
で表わされる含フッ素アクリレートを示す。)



(式中、Ph はフェニレン基、R'' は炭素原子数 5~15 のパーフルオロアルキル基を示す。)

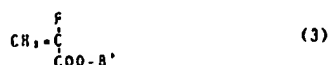
で表わされる基である。

含フッ素重合体に含フッ素アクリレート (1) 以外に含有させることができる単量体としては、例えば式：



(式中、A は水素原子、塩素原子またはメチル基、B は炭素原子数 1~10 のアルキル基、炭素原子数 6~8 の脂環式基または炭素原子数 1~10 のフルオロアルキル基を示す。)

で表わされる単量体、式：



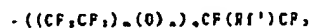
(式中、B' は炭素原子数 1~10 のアルキル基または炭素原子数 6~8 の脂環式基を示す。)

で表わされる単量体、エチレン、プロピレン、スチレンさらにはビニル基、ヒドロキシル基、カル

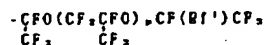
10 重量% 含む含フッ素重合体からなる撥水撥油剤である。

含フッ素重合体の数平均分子量 (ゲルパーミエーションクロマトグラフィーによる) は、1 万~400 万の範囲、固有粘度 (η) (溶媒：メタキシレンヘキサフルオライド、メチルエチルケトン、クロロホルム、1,1,1-トリクロロエタン等、温度：35℃) でいうと、0.25~3.0 の範囲が好ましい。分子量が小さすぎると被処理物品より剥がれやすく、膜強度も小さい。大きすぎると被処理物品に塗布し難くなる。

前記 R' 基は、重合体の撥水撥油性の上から、好ましくは式：



(式中、n は 1~5 の整数、n は 0 または 1、q は 1~5 の整数、R' はフッ素原子またはトリフルオロメチル基を示す。)、式：



(式中、p は 0 または 1~5 の整数、R' は前記と同じ。) または式：

ボキシシル基、グリシジル基、ジアルキルアミノ基またはトリアルコキシシリル基等の官能基を有するアクリレートまたはメタアクリレート等のエチレン性不飽和単量体を挙げることができる。

含フッ素アクリレート (1) の例としては、 $\text{CH}_2=\text{CF}-\text{COOCH}_2\text{CH}_2\text{C}_6\text{F}_5$ 、 $\text{CH}_2=\text{CF}-\text{COOCH}_2\text{C}_6\text{F}_5$ 、 $\text{CH}_2=\text{CF}-\text{COOCH}_2\text{C}_6\text{F}_4$ 、 $\text{CF}(\text{CF}_3)_2$ 、 $\text{CH}_2=\text{CF}-\text{COOCH}_2-\text{CF}(\text{CF}_3)\text{OCF}_2\text{CF}(\text{CF}_3)\text{OC}_2\text{F}_5$ 、 $\text{CH}_2=\text{CF}-\text{COOCH}_2-\text{CF}(\text{CF}_3)\text{OC}_2\text{F}_5$ 、 $\text{CH}_2=\text{CF}-\text{COOCH}_2\text{CH}_2-\text{N}(\text{CH}_3)\text{SO}_2\text{C}_6\text{F}_5$ 、 $\text{CH}_2=\text{CF}-\text{COOCH}_2\text{CH}(\text{OH})\text{CH}_2\text{C}_6\text{F}_5$ 、 $\text{CH}_2=\text{CCl}-\text{COOCH}_2\text{CH}_2\text{C}_6\text{F}_5$ 、 $\text{CH}_2=\text{CCl}-\text{COOCH}_2\text{CF}(\text{CF}_3)\text{OC}_2\text{F}_5$ 、 $\text{CH}_2=\text{C}(\text{CF}_3)-\text{COOCH}_2\text{CH}_2\text{C}_6\text{F}_5$ 等を挙げることができる。

単量体 (2) の例としては、 $\text{CH}_2=\text{CHCOOCH}_3$ 、 $\text{CH}_2=\text{CHCOOC}_2\text{H}_5$ 、 $\text{CH}_2=\text{CHCOO-R}'$ (但し、R' はシクロヘキシル基である。)、 $\text{CH}_2=\text{C}(\text{CH}_3)\text{COOCH}_3$ 、 $\text{C}_6\text{H}_5-\text{C}(\text{CH}_3)\text{COOC}_2\text{H}_5$ 、 $\text{CH}_2=\text{C}(\text{CH}_3)\text{COOCH}_2\text{CH}_2\text{C}_6\text{F}_5$ 、 $\text{CH}_2=\text{C}(\text{Cl})\text{COOCH}_3$ 等を挙げることができる。

単量体 (3) の例としては、 $\text{CH}_2=\text{CF}-\text{COOCH}_3$ 、 $\text{CH}_2=\text{CF}-\text{COO-R}'$ (但し、R' は前記と同じ。)、 $\text{CH}_2=\text{CF}-\text{COOC}_2\text{H}_5$ 等を挙げることができる。

官能基を有するアクリレートまたはメタアクリレートの例としては、 $\text{CH}_2=\text{C}(\text{CH}_3)\text{COO}(\text{CH}_2\text{CH}_2\text{O})_n$ 、 $\text{COC}(\text{CH}_3)=\text{CH}_2$ 、 $\text{CH}_2=\text{C}(\text{CH}_3)\text{COO}(\text{CH}_2)_n$ 、 $\text{COC}(\text{CH}_3)=\text{CH}_2$ 、 $\text{CH}_2=\text{C}(\text{CH}_3)\text{COCCH}_2\text{CH}(\text{OCOC}(\text{CH}_3)=\text{CH}_2)\text{CH}_2\text{OCOC}(\text{CH}_3)=\text{CH}_2$ 、 $\text{CH}_2=\text{C}(\text{CH}_3)\text{COCCH}_2\text{CH}_2\text{OH}$ 、 $\text{CH}_2=\text{CHCOO}-\text{R}^*$ (但し、 R^* はグリシジル基である。)、 $\text{CH}_2=\text{C}(\text{CH}_3)\text{COOCH}_2\text{CH}_2\text{CH}_2\text{Si}(\text{OCH}_3)_3$ 等を挙げることができる。

含フッ素アクリレート(1)を10重量%以上含有する含フッ素重合体、特に前記Xがフッ素原子または塩素原子のアクリレートを含む重合体からなる薄膜は、強靱で良好な可視性を有し、被処理物品に対する接着性がよい。

単量体(2)としてのエチレン、プロピレン、スチレン等の安価な単量体は、含フッ素重合体のコストを下げるのに有効であり、機能上は含フッ素重合体に硬度等を与える効果を有する。単量体(2)としてのエチレン、プロピレン、スチレン等の使用量は、通常90重量%以下である。

含フッ素重合体が官能基を含んでいると、含フ

ともできる。

塊状重合で調製した重合体は、乾燥後溶液にして使用することができる。

溶液重合および塊状重合で使用することができる重合開始剤としては、例えばアゾビスイソブチロニトリル等のアゾ系化合物、ベンゾイルパーオキサイド等のパーオキサイド系化合物等を挙げることができる。

溶液重合および塊状重合では、連鎖移動剤として、ラウリルメルカプタン、チオフェノール等のメルカプタン類を使用することができる。

重合温度は、前記いずれの方法でも、30~100℃が好ましい。

溶液重合または塊状重合で調製した含フッ素重合体は、通常該含フッ素重合体をよく溶解することができる溶解溶媒に溶解した後、溶解含フッ素重合体を析出させない程度の溶解能を有する希釈溶媒で希釈し、被処理物品に適用する。適用方法は、通常の防水潤滑剤と同様、ディップ、はけ塗り、スプレー法等である。濃度は、はけ塗り法で

フッ素重合体の被処理物品に対する接着性が向上する。また、この官能基を利用して含フッ素重合体を架橋することができる。架橋方法は、本技術分野で通常採用されている方法を利用することができる(例えば、特公昭47-42880号公報参照)。含フッ素重合体の官能基の元になる官能基を有するアクリレートまたはメタアクリレートの使用量は、通常30重量%以下である。

本発明の前記含フッ素重合体は、ラジカル重合(溶液、塊状、乳化等)またはアニオン重合で製造することができる。

溶液重合で使用することができる溶媒の例としては、メタキシレンヘキサフルオライド、トリクロロトリフルオロエタン等のフッ素系溶媒、1,1,1-トリクロロエタン等の塩素系溶媒、酢酸エチル、メチルイソブチルケトン、アセトン、トルエン、キシレン等の炭化水素系溶媒等を挙げることができる。溶液重合で調製した重合体は、溶媒から分離・乾燥後改めて溶液にして使用することができる他、重合終了後溶液を単に希釈して使用するこ

は0.1~30重量%、スプレー法では0.05~2重量%程度が好ましい。物品に塗布した後は室温~150℃で乾燥する。

溶解溶媒の例としては、メタキシレンヘキサフルオライド、トリクロロトリフルオロエタン等のフッ素系溶媒、トリクロロエタン等の塩素系溶媒等を挙げることができる。希釈溶媒の例としては、テトラクロロエチレン、トリクロロエチレン等の塩素系溶媒、アセトン等のケトン系溶媒、酢酸エチル等のエステル系溶媒、トルエン等の芳香族系溶媒、n-ペンタン等の飽和脂肪族系溶媒等を挙げることができる。溶解溶媒を希釈溶媒として使用することもできる。

乳化重合で使用する乳化剤としては、ノニオン系の化合物が好ましい。カチオン系の乳化剤も使用可能である。

乳化重合で使用するすることができる重合開始剤としては、水溶性の化合物が好ましく、例えばアゾビスイソブチロアミジン塩酸塩等のアゾ系化合物、コハク酸パーオキサイド等のパーオキサイド系化

合物等を挙げるができる。

重合温度は、30～100℃が好ましい。

乳化重合で調製した含フッ素共重合体は、水性タイプの防水剤として使用することができる。乳化剤は、通常の場合除かなくてもよい。水性タイプの防水剤は、前記方法と同じ方法で適用することができる。水性タイプの防水剤は、水を含んでいるので、乾燥する時は100～150℃に加熱するのが好ましい。

(1) 式のⅠがトリフルオロメチル基の含フッ素アクリレートを単独重合する場合は、重合速度の面でアニオン重合が好ましい。

アニオン重合で使用することができる重合開始剤としては、例えばアルカリ金属、金属水素化物、ナトリウムアミド、グリニヤール試薬、金属アルキル、ビリジン等を挙げることができる。

アニオン重合で使用することができる溶媒としては、トルエン等の芳香族系溶媒、テトラヒドロフラン等のエーテル系溶媒等を挙げるができる。

(実施例)

実施例 1

200cc のガラス製アンプルに式： $\text{CH}_2=\text{CF}-\text{COOC}$
 $\text{H}_2\text{CF}(\text{CF}_3)\text{OC}_2\text{F}_5$ で表わされる単量体（以下、 αPF5
 FO という。）50g、グリシジルメタクリレート
（以下、GMA という。）4g、メタキシレンヘキサフルオライド（以下、 $\alpha\text{-XHF}$ という。）80g およびアゾビスイソブチロニトリル 0.5g を入れ、メタノール/ドライアイスを使用してフリーザー（freeze-thaw）法で脱気・空気を三回繰り返したあと密封した。

アンプルを50℃の恒温槽に30時間浸漬した。

その後、反応混合物を石油エーテル中にあげ、沈澱した含フッ素重合体を乾燥した。52g の含フッ素重合体を得た。

溶媒として $\alpha\text{-XHF}$ を使用し、温度35℃で測定した該重合体の（ η ）は、1.12であった。

元素分析の結果は、炭素 30.2 %およびフッ素 54.4%で、前記単量体のほぼ全てが重合していることがわかった。

アニオン重合の重合は、通常 $1 \times 10^{-4} \sim 10^{-3}$ 程度の高真空下あるいは乾燥不活性ガス雰囲気下で行う。重合温度は、通常 -100 ～ 70℃である。

アニオン重合で製造した重合体は、前記溶液重合で製造した重合体と同様の方法で被処理物品に適用することができる。

本発明の防水剤は、耐摩損性の要求される用途、例えばテント、シートカバー、傘、レインコート、靴、帽子、袍、ジャケット、ジャンパー、エプロン、ブレザー、スラックス、スカート、着物、カーペット、ソファー、カーテン等の各種固体物質に防水剤性を付与するための処理に使用することができる。

得られた重合体を30重量%の $\alpha\text{-XHF}$ （溶解溶媒）溶液にした後、この溶液をトリクロロトリフルオロエタン（希釈溶媒）でさらに 0.5重量%まで希釈した。

前記希釈液を厚さ3mmのポリウレタン被覆不織布からなる合成皮革（デュポン社製コルファム）上に刷毛で塗布した後80℃で30分間加熱し、接着性試験試料を作成した。

該試料の作成直後と10,000回 120°屈伸動作を行った後の水および α -ヘキサデカンとの接触角を測定した。結果を第2表に示す。

実施例 2～9 および比較例 1～3

単量体、重合体溶解溶媒および希釈溶媒として第1表に示すものを使用し、実施例1と同様の操作で接着性試験試料を作成した。試験結果を第2表に示す。

第 1 表

	単量体と組成比 (重量)	(η)	溶解溶媒	希釈溶媒
実施例 2	α F6FO/ MA/GMA = 66/30/4	0.98	n-XHP	CH_2Cl_2
実施例 3	α F6FO/ MA/GMA = 50/46/4	0.90	-	-
実施例 4	α F6FO/ MA/GMA = 28/77/5	0.91	-	-
実施例 5	α F17F/ CMS-70/ 30	0.58	$\text{CCl}_2\text{F}-\text{CCl}_2\text{F}$	$\text{CCl}_2\text{F}-\text{CCl}_2\text{F}$
実施例 6	α F17F/ GMA-90/ 10	0.62	n-XHP	-
実施例 7	α F17F/ SA/17FA/ GMA-50/ 20/25/5	0.71	-	-
実施例 8	α F119F/ MA/GMA = 50/45/5	0.85	-	-
実施例 9	α F119F/ EGMA/MA /SHA-25/ 2/58/15	0.41	CH_2Cl_2	-

第 1 表 (続き)

	単量体と組成比 (重量)	(η)	溶解溶媒	希釈溶媒
実施例 10	α C17F/ SA/GMA = 60/35/5	1.25	n-XHP	CH_2Cl_2
実施例 11	α C17F/ MA/GMA = 70/25/5	1.30	-	$\text{CCl}_2\text{F}-\text{CCl}_2\text{F}$
実施例 12	α C1119F/ LA/GMA = 50/45/5	1.60	-	CH_2Cl_2
比較例 1	17FMA/SA /GMA = 50/45/5	0.35	CH_2Cl_2	$\text{CCl}_2\text{F}-\text{CCl}_2\text{F}$
比較例 2	17FA/GMA -90/10	0.32	n-XHP	-
比較例 3	19PA/MA/ GMA =65/ 30/5	0.68	-	-

第 1 表において、単量体を示す各略号は、次の単量体を意味する。以下、同意義。

MA : $\text{CH}_2=\text{CHCOOCH}_3$,

α F17F : $\text{CH}_2=\text{CFCOOCH}_2\text{CH}_2(\text{CF}_2\text{CF}_2)_3\text{CF}_2\text{CF}_3$,

CMS : $\text{CH}_2=\text{CH}-\text{Ph}-\text{CH}_2\text{Cl}$ (但し、Phはフェニル基である。)

レン基である。)

SA : $\text{CH}_2=\text{CHCOOCH}_2\text{CH}_3$,

17FA : $\text{CH}_2=\text{CHCOOCH}_2\text{CH}_2\text{C}_6\text{F}_5$,

α F119F : $\text{CH}_2=\text{CFCOOCH}_2\text{CH}_2(\text{CF}_2\text{CF}_2)_3\text{CF}(\text{CF}_3)_2$,

EGMA : $\text{CH}_2=\text{C}(\text{CH}_3)\text{COO}(\text{CH}_2\text{CH}_2\text{O})_2\text{COC}(\text{CH}_3)=\text{CH}_2$,

SHA : $\text{CH}_2=\text{C}(\text{CH}_3)\text{COOCH}_2\text{CH}_3$,

17FMA : $\text{CH}_2=\text{C}(\text{CH}_3)\text{COOCH}_2\text{CH}_2\text{C}_6\text{F}_5$,

19FA : $\text{CH}_2=\text{CHCOOCH}_2\text{CH}_2\text{C}_6\text{F}_5$,

α C117F : $\text{CH}_2=\text{CClCOOCH}_2\text{CH}_2(\text{CF}_2\text{CF}_2)_3\text{CF}_2\text{CF}_3$,

α C1119F : $\text{CH}_2=\text{CClCOOCH}_2\text{CH}_2(\text{CF}_2\text{CF}_2)_3\text{CF}(\text{CF}_3)_2$,

LA : $\text{CH}_2=\text{CHCOOCH}_2\text{CH}_3$,

第 2 表

	接 触 角 ($^\circ$) 作成直後/屈伸操作後	
	水	n-ヘキサデカン
実施例 1	110/108	74/52
" 2	111/105	74/56
" 3	120/101	71/50
" 4	116/100	66/48
" 5	123/110	80/58
" 6	122/115	80/52
" 7	120/105	78/49
" 8	108/102	75/50
" 9	110/100	70/45
" 10	118/105	76/49
" 11	120/108	78/50
" 12	113/102	75/50
比較例 1	102/73.6	68/15
" 2	108/70	69/20
" 3	106/71	69/19

実施例13および比較例4

実施例1と比較例3で調製した重合体と同じ重合体をそれぞれ5重量%になるように m -XHFに溶解した後、直径9cmのシャーレ上にキャストし、乾燥した。厚み約82 μ mのシートを得た。

これらシートの破断強度とその時の伸び率を調べたところ、以下の通りであった。

実施例1の重合体（実施例13に当たる）：

破断強度 = 1.0 kgf/mm²

伸び率 = 300%

比較例3の重合体（比較例4に当たる）：

破断強度 = 0.26 kgf/mm²

伸び率 = 450%

実施例14

攪拌機、温度計、還流器および滴下ロートを備えた3Lの四つ口フラスコに水 1.9L、アセトン 400g、 α -F6FD 300g、MA 19g、EGMA 1gおよび乳化剤（日本油脂製 M-220）40gを入れ、系内に窒素を吹き込み酸素を除いた。65℃の恒温槽に入れ、温度が一定になったところで、アゾビス

イソブチロアミジン・塩酸塩 1.6gを溶解した水 0.1Lを滴下し、重合を開始した。4時間後、固形分12重量%のディスパージョンを得た。一部をサンプリングして単量体組成と (η) を求めた。

単量体組成（重量%）： α -F6FD/MA/EGMA=93.7/5/0.3（元素分析：炭素 39.6%およびフッ素 55.0%）、 (η) = 0.68。

前記得られたディスパージョンをバッディング槽中で 0.5重量%になるように水で希釈した。ポリエステル製布をバッディング浴に浸漬し、絞って水を切った後、80℃で3分間乾燥し、150℃で3分間熱処理して試験試料を作成した。

この試料について、JIS L 1006の撥水性試験とAATCC 118-1966Tの撥油性試験を行ったところ、各々100°とNo.6の結果を得た。

同じ試料を家庭用電気洗濯機を使用し、浴比 1:50、洗剤 ザブ、温度40℃の条件で洗濯した後、風乾し、140℃のアイロンをかけるかけ、再び前記両試験を行ったところ、各々100°とNo.5の結果を得た。

比較例5

単量体を 17FA 300g、MA 19gおよびEGMA 1gに変更した他は、実施例14と同様の条件で重合および試験試料の作成を行った。重合体の (η) は、0.38であった。

実施例14と同様の条件で洗濯前後の撥水性試験と撥油性試験を行ったところ、撥水性は100°から70°へ、撥油性はNo.3からNo.0へ低下していた。

（発明の効果）

本発明の撥水撥油剤は、 α 位にフッ素原子、塩素原子またはフッ素原子含有基を有する含フッ素アクリレートを構成成分とする重合体からなるものである。膜強度や被処理物品に対する接着性等が従来の撥水撥油剤に比べて優れており、洗濯等に対して耐久性を有している。

以上

特許出願人 ダイキン工業株式会社

INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP02/03686

A. CLASSIFICATION OF SUBJECT MATTER

Int.Cl⁷ C09K3/18, C09D157/08, 133/16

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

Int.Cl⁷ C09K3/18, C09D157/08, 133/16

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Jitsuyo Shinan Koho	1922-1996	Toroku Jitsuyo Shinan Koho	1994-2002
Kokai Jitsuyo Shinan Koho	1971-2002	Jitsuyo Shinan Toroku Koho	1996-2002

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	JP 63-75082 A (Daikin Industries, Ltd.), 05 April, 1988 (05.04.88), Pages 2, upper right column, line 4 to lower right column, line 4; page 2, lower right column, line 10; examples 3 to 4 (Family: none)	1-4
Y	JP 1-153784 A (Daikin Industries, Ltd.), 15 June, 1989 (15.06.89), Claim 1; page 2, lower left column, line 14 to page 3, upper left column, line 15; page 3, upper right column, lines 11 to 20; page 3, lower right column, line 20 to page 4, upper right column, line 11; examples 1 to 8 (Family: none)	1-4

☒ Further documents are listed in the continuation of Box C.
 ☐ See patent family annex.

* Special categories of cited documents:	"I" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"A" document defining the general state of the art which is not considered to be of particular relevance	"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"E" earlier document but published on or after the international filing date	"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	"&" document member of the same patent family
"O" document referring to an oral disclosure, use, exhibition or other means	
"P" document published prior to the international filing date but later than the priority date claimed	

Date of the actual completion of the international search
19 June, 2002 (19.06.02)Date of mailing of the international search report
02 July, 2002 (02.07.02)Name and mailing address of the ISA/
Japanese Patent Office

Authorized officer

Telephone No.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP02/03686

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	EP 247489 A2 (Daikin Industries, Ltd.), 02 December, 1987 (02.12.87), Page 1, line 23 to page 4, line 22; examples 1 to 4 & JP 63-99285 A Page 2, upper left column, line 6 to lower right column, line 12; examples 1 to 4	1-4
Y	JP 2000-160147 A (Asahi Glass Co., Ltd.), 13 June, 2000 (13.06.00), Par. No. [0017] (Family: none)	1-4

PATENT COOPERATION TREATY

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INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference 1320/7-PCT	FOR FURTHER ACTION See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/EP99/05205	International filing date (day/month/year) 21/07/1999	Priority date (day/month/year) 22/07/1998
International Patent Classification (IPC) or national classification and IPC C08F2/22		
Applicant ELOTEX AG et al.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.



2. This REPORT consists of a total of 5 sheets, including this cover sheet.

- ☐ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of 3 sheets.

3. This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☐ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☐ Certain defects in the international application
- VIII ☒ Certain observations on the international application

Date of submission of the demand 16/02/2000	Date of completion of this report 19. 02. 00
Name and mailing address of the international preliminary examining authority:  European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer Rouault, Y Telephone No. +49 89 2399 8524 

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/EP99/05205

I. Basis of the report

1. This report has been drawn on the basis of (*substitute sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments.*):

Description, pages:

1-8,10-13,15-24	as originally filed			
9,14	as received on	04/09/2000	with letter of	30/08/2000

Claims, No.:

1-15,17-29	as originally filed			
16	as received on	04/09/2000	with letter of	30/08/2000

Drawings, sheets:

1/2,2/2	as originally filed
---------	---------------------

2. The amendments have resulted in the cancellation of:

- ☐ the description, pages:
- ☐ the claims, Nos.:
- ☐ the drawings, sheets:

3. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

4. Additional observations, if necessary:

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/EP99/05205

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes:	Claims	1-26
	No:	Claims	
Inventive step (IS)	Yes:	Claims	
	No:	Claims	16-26
Industrial applicability (IA)	Yes:	Claims	1-26
	No:	Claims	

2. Citations and explanations

see separate sheet

VIII. Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

see separate sheet

Re Item V

Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Reference is made to the following document:

D1: EP-A-696602

Novelty

The claims 1-15 are novel (Art. 33(2) PCT). None of the documents cited in the search report describe a process as defined in the independent claim 1.

The claims 16-29 are novel (Art. 33(2) PCT). None of the documents cited in the search report describe a product as defined in claim 16.

2. **Inventiveness of claims 1-16**

The claims 1-15 are based on an inventive step (Art 33(3) PCT).

The present application describes an alternative process for the production of encapsulated hydrophilic polymers to the one proposed in D1 with the advantage that the products are made in a one-step-process wherein no separation between the production of the core and the production of the shell is necessary.

The cited documents, even if combined, could not lead to the present process.

3. **Inventiveness of claims 16-26**

- 3.1 The disclaimer in claim 16 renders the claim new. However, the technical effect obtained by the restriction is not clear. Example 3 of the present application is out of the disclaimed range only because the amount of acid-functional monoethylenically unsaturated monomers is larger than 10 wt%. There is apparently no technical effect bound with this feature.

- 3.2 Example 2, which is presented as a comparative example, is in fact an embodiment of the invention according to claims 6 and 11.

The non water soluble methyl methacrylate, which has a $T_g > 30^\circ\text{C}$, is used in

place of styrene. As a consequence:

- Example 2 is not a comparative example and a comparison with the prior art is not given by example 2.
- it is stated in example 2 that the raspberry-like structure is not obtained with MMA, so that it is not possible to obtain a different morphology as the encapsulated morphology in all cases. The possibility to obtain different latex morphology is therefore not confirmed.

3.3 Some alleged advantages, like the processability, are not clearly defined and seem to be subjective, so that a technical effect is not clearly defined.

3.4 Hence, from the description and the examples, it is not possible to say which technical effect is achieved for the product in the present application, in particular by the use of a semicontinuous instead of a continuous polymerisation that seems to be the distinguishing feature between the present application and the prior art (Art. 33(3) PCT).

The present claims 16-26 are apparently not based on an inventive step (Art 33(3) PCT).

The industrial applicability (Art 33(4) PCT) is obvious.

Re Item VIII

Certain observations on the international application

It is clear from the description on page 11 l. 14-24 that the hydrophilic, ethylenically unsaturated monomer in claim 1 b) has to contain at least one acid functionality. This feature is essential to the definition of the invention.

Since independent claim 1 does not contain this feature, it does not meet the requirement following from Article 6 PCT taken in combination with Rule 6.3(b) PCT that any independent claim must contain all the technical features essential to the definition of the invention.

saturated (co)monomers, and at least one hydrophilic, ethylenically unsaturated (co)monomer in a quantity of about 5 to 30 wt. %, based on the total weight of the ethylenically unsaturated (co)monomers.

5 It is important for the solution of the set problem, that the above-indicated parameters are respected. In particular, the glass transition temperature of the non-ionic monomer must be above about 30°C. Preferably, the glass transition temperature of the nonionic monomer is between about 30 and 120°C, and in particularly preferred manner between about 50 and 110°C. This permits the setting of a high glass transition temperature of the polymer in the outer phase (shell), which contributes to the obtaining of a homogeneous distribution of the reactive groups present in the encapsulated, inner phase (core). On ~~exceeding~~ ^{falling down} this Tg value, it is no longer possible to ensure a homogeneous distribution, particularly in the case of a large number of reactive groups.

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25
The setting of the glass transition temperature Tg takes place in known manner by the choice and quantity of the monomers used. The weight fractions of the possible comonomers are chosen in such a way that the glass transition temperature Tg (midpoint temperature according to ASTM D3418-82) of the film formation of the redispersible particles produced leads to the desired, modifying action. The glass transition temperature can e.g. be measured by DSC methods or determined theoretically by calculations. In the present invention, the glass transition temperatures are calculated according to the Fox trial and error method (T.G. Fox, Bull. Am. Phy. Soc. (ser II) 1, 123 (1956) and Ullmann's Enzyklopädie der Technischen Chemie, vol. 19, 4th edition, Verlag Chemie, Weinheim, 1980, pp 17/18). Thus, for the glass transition temperature applies:

$$\frac{1}{T_g} = \frac{w_A}{T_{gA}} + \frac{w_B}{T_{gB}} + \dots + \frac{w_n}{T_{gn}}$$

swelling of the polymer particles. Suitable swelling agents include bases of all types.

5 The planned control of the characteristics profile of the substrates to be modified, i.e. the improved action by the added particles, is more effective in proportion to the ^{fineness}~~fineness~~ of the particles, i.e. it is particularly advantageous of the dispersed polymerizate particles have a particularly small diameter. As a result of the process control according to the invention, it is possible to produce in planned manner substantially monodisperse latex particles with corresponding particle diameters. In this context "monodisperse" means that the average particle diameter varies by about $\pm 10\%$. The average diameter of the latex particles is in a range of about 30 to 1000 and in particular about 50 to 600 nm.

10
15 The invention also relates to aqueous dispersions of latex particles having a heterogeneous morphology, obtainable by the above described process. According to a preferred embodiment of the invention the dispersion can comprise an aqueous dispersion 1 with one kind of latex particles and a further aqueous dispersion 2 with other latex particles. The weight ratio of dispersion 1 to dispersion 2 is preferably in the range of about 5:95 to 95:5, especially about 10:90 to 90:10, particularly about 20:80 to 80:20. Dispersion 2 can comprise an aqueous dispersion of homopolymers or copolymers selected from the group consisting of the monomers vinyl acetate, ethylene, vinyl versatate, acrylate, methacrylate, styrene and/or butadiene. This is only an exemplary listing and as a matter of course those skilled in the art know further monomers which can be used. By
20
25 adding a further dispersion the properties of the dispersion according to the invention can be optimized accordingly.

30 The invention also relates to latex particles having a heterogeneous morphology, which are obtainable from the aqueous dispersion by corresponding removal of the water. The latex particles obtainable according to the invention have a het-

HAGEMANN, BRAUN & HELD

PATENTANWÄLTE

EUROPEAN PATENT ATTORNEYS

MÜNCHEN · HANNOVER

PCT/EP 99/05205
Applicant: Elotex AG
Our Ref.: Pat 1320/7-99-PCT

München, den 30.08.00
Dr.H/hn (cp)

Novel Claim 16

16. Aqueous dispersion of latex particles having a heterogeneous morphology, obtainable according to a process according to at least one of the claims 1 to 15, wherein 0.1 % by weight to 10 % by weight, based on the total weight of the shell polymer, of an acid-functional monoethylenically unsaturated monomer is excluded in case the stabilizer used has no cationic functionality.

* * *

AMENDED SHEET

ART 34 Amdt

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WORLD INTELLECTUAL PROPERTY ORGANIZATION
International Bureau



INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

<p>(51) International Patent Classification ⁷ : C08F 2/22, 291/00, C04B 24/26</p>	<p>A1</p>	<p>(11) International Publication Number: WO 00/05276 (43) International Publication Date: 3 February 2000 (03.02.00)</p>
<p>(21) International Application Number: PCT/EP99/05205 (22) International Filing Date: 21 July 1999 (21.07.99) (30) Priority Data: 198 33 061.8 22 July 1998 (22.07.98) DE (71) Applicant (for all designated States except US): ELOTEX AG [CH/CH]; Industriestrasse 17a, CH-6203 Sempach Station (CH). (72) Inventors; and (75) Inventors/Applicants (for US only): WILLIMANN, Hongli [CH/CH]; Burgstrasse 2, CH-5634 Merenschwand (CH). KOELLIKER, Robert [CH/CH]; Unterhofstrasse 14, CH-6208 Oberkirch (CH). (74) Agents: HAGEMANN, Heinrich et al.; Hagemann, Braun & Held, Postfach 860 329, D-81630 München (DE).</p>		<p>(81) Designated States: AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZA, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).</p> <p>Published <i>With international search report.</i> <i>Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i></p>
<p>(54) Title: A PROCESS FOR THE PREPARATION OF AQUEOUS DISPERSIONS OF LATEX PARTICLES HAVING A HETEROGENEOUS MORPHOLOGY, THE LATEX PARTICLES OBTAINABLE WITH THE PROCESS; THE DISPERSIONS AND REDISPERSIBLE POWDERS, AS WELL AS THE USE THEREOF</p> <p>(57) Abstract</p> <p>The invention relates to a process for the preparation of aqueous dispersions of latex particles having a heterogeneous morphology by a semicontinuous emulsion polymerization, comprising the emulsion polymerizing of ethylenically unsaturated (co)monomers, accompanied by the addition of cationic and/or anionic and/or nonionic emulsifiers and/or protective colloids as stabilizers, which are directly used as such or synthesized in situ, the semicontinuous emulsion polymerization being performed in the presence of the stabilizer or stabilizers with a monomer mixture, which a) contains at least one nonionic, ethylenically unsaturated monomer with a glass transition temperature T_g above about 30 °C in a quantity of about 10 to 70 wt.%, based on the total weight of ethylenically unsaturated (co)monomers and b) at least one hydrophilic, ethylenically unsaturated monomer in a quantity of about 5 to 30 wt.%, based on the total weight of ethylenically unsaturated (co)monomers.</p>		

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BB	Barbados	GH	Ghana	MG	Madagascar	TJ	Tajikistan
BE	Belgium	GN	Guinea	MK	The former Yugoslav Republic of Macedonia	TM	Turkmenistan
BF	Burkina Faso	GR	Greece			TR	Turkey
BG	Bulgaria	HU	Hungary	ML	Mali	TT	Trinidad and Tobago
BJ	Benin	IE	Ireland	MN	Mongolia	UA	Ukraine
BR	Brazil	IL	Israel	MR	Mauritania	UG	Uganda
BY	Belarus	IS	Iceland	MW	Malawi	US	United States of America
CA	Canada	IT	Italy	MX	Mexico	UZ	Uzbekistan
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CG	Congo	KE	Kenya	NL	Netherlands	YU	Yugoslavia
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CI	Côte d'Ivoire	KP	Democratic People's Republic of Korea	NZ	New Zealand		
CM	Cameroon			PL	Poland		
CN	China	KR	Republic of Korea	PT	Portugal		
CU	Cuba	KZ	Kazakstan	RO	Romania		
CZ	Czech Republic	LC	Saint Lucia	RU	Russian Federation		
DE	Germany	LI	Liechtenstein	SD	Sudan		
DK	Denmark	LK	Sri Lanka	SE	Sweden		
EE	Estonia	LR	Liberia	SG	Singapore		

INTERNATIONAL SEARCH REPORT

International Application No

PC./EP 99/05205

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C08F2/22 C08F291/00 C04B24/26

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C08F C04B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 0 426 391 A (MITSUI TOATSU CHEMICALS INC.) 8 May 1991 (1991-05-08) cited in the application ---	
A	EP 0 696 602 A (ROHM AND HAAS CO.) 14 February 1996 (1996-02-14) cited in the application -----	

☐

Further documents are listed in the continuation of box C.

☒

Patent family members are listed in annex.

* Special categories of cited documents :

- "A" document defining the general state of the art which is not considered to be of particular relevance
- "E" earlier document but published on or after the international filing date
- "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- "O" document referring to an oral disclosure, use, exhibition or other means
- "P" document published prior to the international filing date but later than the priority date claimed

- "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- "&" document member of the same patent family

Date of the actual completion of the international search

12 November 1999

Date of mailing of the international search report

24/11/1999

Name and mailing address of the ISA

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Cauwenberg, C

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/EP 99/05205

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
EP 426391 A	08-05-1991	JP 2872710 B	24-03-1999
		JP 3140306 A	14-06-1991
		AU 623557 B	14-05-1992
		AU 6494090 A	02-05-1991
		DE 69028679 D	31-10-1996
		DE 69028679 T	22-05-1997
		US 5216044 A	01-06-1993
EP 696602 A	14-02-1996	US 5494971 A	27-02-1996
		AU 706410 B	17-06-1999
		AU 2842395 A	22-02-1996
		BR 9503621 A	30-04-1996
		CA 2155808 A	13-02-1996
		CN 1130634 A	11-09-1996
		FI 953718 A	13-02-1996
		JP 8081506 A	26-03-1996
		SG 47351 A	17-04-1998
		US 5545695 A	13-08-1996

PATENT COOPERATION TREATY

PCT

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference 1320/7-PCT	FOR FURTHER ACTION see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.	
International application No. PCT/EP 99/ 05205	International filing date (day/month/year) 21/07/1999	(Earliest) Priority Date (day/month/year) 22/07/1998
Applicant ELOTEC AG et al.		

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 2 sheets.

☒ It is also accompanied by a copy of each prior art document cited in this report.

1. Basis of the report

a. With regard to the language, the International search was carried out on the basis of the International application in the language in which it was filed, unless otherwise indicated under this item.

☐ the International search was carried out on the basis of a translation of the International application furnished to this Authority (Rule 23.1(b)).

b. With regard to any nucleotide and/or amino acid sequence disclosed in the International application, the International search was carried out on the basis of the sequence listing:

☐ contained in the International application in written form.

☐ filed together with the International application in computer readable form.

☐ furnished subsequently to this Authority in written form.

☐ furnished subsequently to this Authority in computer readable form.

☐ the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the International application as filed has been furnished.

☐ the statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished

2. ☐ Certain claims were found unsearchable (See Box I).

3. ☐ Unity of invention is lacking (see Box II).

4. With regard to the title,

☒ the text is approved as submitted by the applicant.

☐ the text has been established by this Authority to read as follows:

5. With regard to the abstract,

☒ the text is approved as submitted by the applicant.

☐ the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this International search report, submit comments to this Authority.

6. The figure of the drawings to be published with the abstract is Figure No.

☐ as suggested by the applicant.

☐ because the applicant failed to suggest a figure.

☐ because this figure better characterizes the invention.

☒ None of the figures.

INTERNATIONAL SEARCH REPORT

International Application No.

CT/EP 99/05205

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Cauwenberg, C

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

T/EP 99/05205

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